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surface ozone trends**

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Modelling future changes in surface ozone: a parameterized approach

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Abstract

This study describes a simple parameterization to estimate regionally averaged changes in surface ozone due to past or future changes in anthropogenic precursor emissions based on results from 14 global chemistry transport models. The method successfully reproduces the results of full simulations with these models. For a given emission scenario it provides the ensemble mean surface ozone change, a regional source attribution for each change, and an estimate of the associated uncertainty represented by the variation between models. Using the Representative Concentration Pathway (RCP) emission scenarios as an example, we show how regional surface ozone is likely to respond to emission changes by 2050 and how changes in precursor emissions and atmospheric methane contribute to this. Surface ozone changes are substantially smaller than expected with the SRES A1B, A2 and B2 scenarios, reflecting the assumptions of more stringent precursor emission controls under the RCP scenarios. We find an average difference of around 5 ppb between the outlying RCP 2.6 and RCP 8.5 scenarios, about 75 % of which can be attributed to differences in methane abundance. The study reveals the increasing importance of limiting atmospheric methane growth as emissions of other precursors are controlled, but highlights differences in modelled ozone responses to methane changes of as much as a factor of two, indicating that this remains a major uncertainty in current models.

1 Introduction

Increases in anthropogenic emissions of ozone precursors are believed to make a substantial contribution to the rising levels of surface ozone (O_3) observed at many long-term measurement stations over past decades (Oltmans et al., 2006). As a strong oxidant detrimental to human health and damaging to plant growth and crop yields, surface O_3 contributes to poor air quality and to economic and environmental damage. Understanding the reasons for its growth presents a considerable challenge, as the

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balance of natural and anthropogenic, regional and global changes contributing to its growth varies greatly over the globe and remains poorly characterized (e.g., Lelieveld and Dentener, 2000; Sudo and Akimoto, 2007). While surface O_3 is often considered a regional pollutant that can be addressed with regional-scale precursor emission controls, it is also a global pollutant that can influence air quality over intercontinental scales (e.g., Akimoto, 2003; TF-HTAP, 2010). It remains unclear how regional emission controls aimed at reducing surface O_3 may be offset by global “background” O_3 increases, by changes in the abundance of longer-lived O_3 precursors such as methane (CH_4) or by changes in chemical processing or transport driven by future shifts in climate (Fiore et al., 2009; Jacob and Winner, 2009; TF-HTAP, 2010). Understanding the anthropogenic contribution to changes in surface O_3 requires a sufficiently good understanding of the chemical and dynamical processes controlling O_3 and the sources and fates of its precursors to explain observed changes, and hence to reduce the current uncertainty in our estimates of future changes in O_3 affecting air quality and climate.

This study explores the contribution of changes in anthropogenic O_3 precursor emissions to changes in the regional and global abundance of surface O_3 . It describes a simple approach to quantify surface O_3 changes based on regional precursor emission changes derived from global chemical transport model simulations from a recent model intercomparison. This is applied to past and future emission scenarios to explore the range of surface O_3 responses expected over different parts of the world and to provide a source attribution for these changes. The approach provides a measure of the uncertainty in the estimated responses as represented by the variation over the 14 independent models contributing to the study. We start by describing and evaluating the parameterization used in Sect. 2–4, and test it with historical emission changes in Sect. 5. We then explore a range of future emission scenarios in Sect. 6, quantifying the uncertainty in the expected O_3 changes and providing a regional source attribution for these changes. We conclude by suggesting how the approach introduced here may be used to inform emission controls targeting air quality.

2 Model simulations and emission scenarios

Under the Convention on Long-range Transboundary Air Pollution (LRTAP), the task force on Hemispheric Transport of Air Pollutants (HTAP) was established to develop a fuller understanding of the transport of a range of key air pollutants over intercontinental scales (TF-HTAP, 2007). A series of multi-model intercomparison experiments was coordinated by HTAP to provide a consistent quantification of intercontinental source-receptor relationships between major industrialised regions. Global and regional models of atmospheric chemistry and transport were run with 2001 meteorological conditions and with best estimates of natural and anthropogenic emissions and a specified atmospheric abundance of CH₄ (1760 ppb), and these provided monthly mean distributions of O₃ and aerosol and their precursors for the year. A simulation with 20 % reduced atmospheric concentrations of CH₄ was performed to determine the impacts of CH₄ abundance on O₃ in each model, and this was followed by a further series of runs with 20 % reductions in anthropogenic emissions of the main O₃ precursors, nitrogen oxides (NO_x), carbon monoxide (CO) and volatile organic compounds (VOCs), individually and together, over each of the four main continental scale regions of interest in turn, see Fig. 1. A response matrix was then generated to quantify the impact of each emission change over each source region on each receptor region in each model, and the resulting source-receptor relationships for O₃ and its precursors are outlined in the HTAP assessment reports (TF-HTAP, 2007, 2010) and explored in further detail elsewhere (Sanderson et al., 2008; Shindell et al., 2008; Fiore et al., 2009).

For the present study, an additional set of four runs were defined with 20 % global emission reductions for each precursor so that the effects of emission changes outside the four HTAP regions, the Rest-of-World response, can be considered. The number of data sets from distinct models or model/meteorology combinations that were contributed for each simulation is summarised in Table 1. Each of these data sets provides the monthly mean spatial distribution of O₃ changes over the global domain for the specified emission change, allowing continental-scale source-receptor relationships to be derived.

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The models contributing to the HTAP intercomparison were run with their own best estimates of emissions for 2001 conditions. Recent studies of future surface O₃ have typically used emission scenarios from the Special Report on Emissions Scenarios (SRES) generated for Intergovernmental Panel on Climate Change (IPCC) assessment reports. Three of the four scenario families introduced (A1, A2 and B2) show large increases in O₃ precursor emissions, and previous assessments of future air quality consequently show very large increases in future O₃ (e.g., Prather et al., 2003). In the present study we contrast these with the new Representative Concentration Pathways (RCP) generated for the Climate Model Intercomparison Project (CMIP5) simulations for the IPCC fifth assessment report along with harmonized historical emissions from 1850 to 2000 from Lamarque et al. (2010). Emissions of O₃ precursors are available from 2000 to 2100 along each RCP pathway, and we use these along with the specified changes in atmospheric CH₄ abundance. The four RCP scenarios represent different assumptions about climate change mitigation measures, and are labelled by the levels of radiative forcing reached by 2100: RCP 2.6 (also referred to as RCP 3-PD), RCP 4.5, RCP 6.0 and RCP 8.5 (van Vuuren et al., 2007; Smith et al., 2006; Fujino et al., 2006; Riahi et al., 2007). Air pollution control policies are assumed to evolve under the different scenarios, and emissions of most O₃ precursors decline by 2050, although more strongly in the cleaner RCP 2.6 scenario than in the high radiative forcing RCP 8.5 scenario, and with substantial differences in regional distribution. We note that the four RCP scenarios represent possible future emission pathways, but have been developed independently and are governed by different assumptions about social, economic and political development.

3 Parameterizing ozone responses

In this study we use model results from the HTAP intercomparison to quantify the impact of a realistic range of changes in anthropogenic precursor emissions on surface O₃ on a global, regional and sub-regional basis. The approach involves scaling surface

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O_3 responses derived from 20 % emission reductions by the fractional emission change for a given emission scenario over each region for each precursor. For each model the HTAP results provide the monthly mean O_3 change, $\Delta O_3(i,j,k)$, over each receptor region, k , for a 20 % reduction in emissions, E_{ij} , of each precursor, i , over each source region, j . The atmospheric CH_4 abundance in these model runs was fixed at 1760 ppb, and so we are not able to explore the effect of CH_4 emission changes; however, we use model runs with 20 % reduced CH_4 abundance (1408 ppb) to determine the regional O_3 response to CH_4 changes, $\Delta O_{3m}(k)$. The monthly mean O_3 response over each region, $\Delta O_3(k)$, is then calculated by summing the individual responses for each of the three precursors (NO_x , CO and VOC) over the five regions encompassing all global sources (N. America, Europe, S. Asia, E. Asia and Rest-of-World) and including the response from the change in global CH_4 abundance:

$$\Delta O_3(k) = \sum_{i=1}^3 \sum_{j=1}^5 f_{ij} \Delta O_3(i,j,k) + f_m \Delta O_{3m}(k) \quad (1)$$

The scale factor for the O_3 response to each regional precursor emission change, f_{ij} , is dependent on the emission scenario and is given by the ratio of the fractional emission change, $\Delta E_{ij}/E_{ij}$, to the 20 % emission change applied in the HTAP simulations:

$$f_{ij} = \frac{\Delta E_{ij}}{0.2 \times E_{ij}} \quad (2)$$

Similarly, the scale factor for the CH_4 response, f_m , is given by the ratio of the global abundance change, ΔCH_4 , to the 20 % abundance change:

$$f_m = \frac{\Delta CH_4}{0.2 \times [CH_4]} \quad (3)$$

These scale factors vary linearly with the size of the applied emission change or change in CH_4 abundance. This simple linear combination of individual regional O_3 responses

works well for small emission changes, and is applicable to larger changes with only minor modifications to account for the small degree of nonlinearity inherent in O_3 chemistry at the continental scales considered here, described below.

A major advantage of this combinatorial approach is its computational simplicity, allowing regional and global O_3 changes to be explored for a wide range of different emission scenarios. Equation (1) can be applied to each atmospheric model in turn to generate a range of O_3 responses that reflect the uncertainties in emissions, chemistry and transport over the contributing models. The approach can be used to determine where the largest uncertainties arise, pinpointing model weaknesses, and to identify emission scenarios that would be of most interest to explore in further detail with full model simulations. In addition, the approach provides an immediate regional source attribution for O_3 changes, something not readily available from full model simulations without a tracer tagging scheme or further sensitivity studies.

4 Testing the approach

To test the validity of the approach and quantify the errors involved, results from the HTAP intercomparison are supplemented here by additional simulations using one of the contributing models, the Frontier Research System for Global Change version of the University of California, Irvine chemical transport model, FRSGC/UCI CTM (Wild et al., 2003).

Ozone responses due to individual precursor emission changes are used here in preference to those due to combined changes in NO_x , CO and VOC emissions as many models also included changes in aerosol in the combined runs. However, changes in tropospheric photochemistry, particularly through the abundance of the hydroxyl radical, OH, couple the effects of these precursor changes such that the O_3 responses are not independent. Over the coarse temporal and spatial scales considered here the effects of individual precursor emission changes are almost linearly additive, as noted in Fiore et al. (2009). Figure 2 shows that the sum of the O_3 responses from each of

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the 20 % precursor emission changes in North American sources closely matches the response from the combined emissions change on a month by month basis, both over the source region itself and over a downwind receptor region. The regional monthly responses from the sum of separate emission changes are marginally larger than those from the combined emission changes, by 2–7 % over the source region and by less than 2 % over receptor regions. Fractional errors are largest over the source region in winter where NO_x emission reductions lead to enhancement in O_3 due to reduced titration by NO , but the absolute O_3 response is small in this season. NO_x emission reductions provide the largest O_3 response but lower the abundance of OH , and hence the O_3 response of CO and VOC reductions is less when all three precursors are reduced together. Larger errors may be expected to occur on smaller spatial scales close to urban source regions which are more greatly influenced by rapid chemical processing, but these effects are not seen at the continental scales considered here, where behaviour is close to linear.

Rest-of-world O_3 responses are required here to account for changes in emissions outside the four HTAP regions. These are derived by subtracting the response due to emissions from each region from that of a global emissions change. To test the validity of this, additional simulations were performed with the FRSGC/UCI CTM applying 20 % anthropogenic precursor emission reductions everywhere outside the HTAP regions for each of the precursors in turn. Figure 2 shows the difference between the sum of the regional and rest-of-world O_3 responses and the response to global emission changes. Over major source regions the response to local emissions dominates and shows a strong seasonality, while the global average response is more uniform. The differences are less than 2 % in all regions except for wintertime in Europe where they reach 4 % due to non-linear responses associated with the greater prevalence of O_3 titration in this region.

The 20 % emission perturbations applied in the HTAP studies were chosen to be small enough to give an approximately linear response while being sufficiently large to provide robust signals in all models. However, the response of O_3 to its precursor

emissions is known to be non-linear (e.g., Lin et al., 1988), and it is important to characterize where these non-linearities become significant. Scaling a 20 % emission reduction by a factor of five has been shown to underestimate the response to a 100 % reduction (Wu et al., 2009), and while this underestimation is relatively small for VOC emissions, generally less than 10 %, it can exceed a factor of two for NO_x emissions (Wu et al., 2009; Grewe et al., 2010), and shows a strong seasonal dependence (Wu et al., 2009). For this reason the sensitivity approach used in the HTAP studies is unsuitable for deriving a full source apportionment for O₃. However, it does not preclude its use in estimating the impact of less severe emission changes.

To determine the limits of the linear scaling, additional simulations were performed with the FRSGC/UCI CTM for NO_x emission changes ranging from complete removal to a doubling of anthropogenic emissions. We focus on emissions from Europe, where deviation from linear behaviour is greatest due to higher latitudes and thus lower insolation which lead to substantial wintertime titration of surface O₃. Figure 3 shows the O₃ responses over the European source region and over a receptor region (N. America) along with the errors associated with linearly scaling a 20 % emission reduction. Non-linear behaviour is clear, particularly over the source region where annual regional mean O₃ is at a maximum in this model, and where additional wintertime titration exceeds summertime production for any further increase in NO_x emissions. However, the nonlinearity is relatively small for small emission changes, and the error in scaling 20 % changes remains below 1 ppb under emission changes of up to ±60 %. In all cases a linear scaling leads to overestimation of O₃, reflecting the curvature of the O₃ response shown in Fig. 3, and indicating that the magnitude of any O₃ change will be underestimated for emission reductions and overestimated for emission increases. Large departures from linearity are typically confined to emission reductions of more than 60 %. Over downwind receptor regions the nonlinear behaviour is weaker, and the fractional error is similar throughout the year so that the largest errors are present in April when the contributions from Europe are greatest. Note that the errors identified here apply to the effects of NO_x emission changes in isolation; simultaneous changes

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in CO and VOC emissions of a similar magnitude reduce the nonlinearity substantially. Complete removal of all anthropogenic emissions over Europe leads to an overestimate of 3 ppb over the source region using the linear scaling compared with the 6 ppb overestimate seen here from removal of NO_x emissions alone.

5 To account for the nonlinearity in O₃ response to larger NO_x emission changes shown in Fig. 3, we replace the scale factor, f , in Eq. (1) with a new factor, g , which has a quadratic dependence on f :

$$g = 0.95f + 0.05f^2 \quad (4)$$

This provides a small amount of curvature equivalent to an incremental reduction of 10% in the O₃ response for each successive 20% emission increase. This simple quadratic approach provides a better fit to the curves for the full CTM simulations shown in Fig. 3, and reduces the errors in the monthly O₃ responses by 20–60% for NO_x emission changes of up to 60%. For emission reductions greater than 60% this correction remains insufficient, and we do not expect the parameterization to work as well under these conditions. The approach is also insufficient in source regions under titration regimes where an emission reduction may lead to an increase in O₃, as seen in January in Fig. 3. Under these conditions we limit the O₃ response by replacing f in Eq. (1) with $2 - g$ for emission reductions, and use the linear scaling f for emission increases. Non-linearity in the O₃ response to CO and VOC emission changes has been shown to be much smaller (Wu et al., 2009), and we therefore retain the linear scale factor, f , for these precursors.

Previous studies have suggested that the O₃ response to CH₄ emissions is approximately linear (Fiore et al., 2008). However, we find that an additional CTM run applying a 20% increase in CH₄ abundance gives an 11% smaller O₃ response over all regions than a run applying a 20% CH₄ decrease. Half of this 11% reduction, about 5%, reflects the feedback of CH₄ on its own lifetime (see, e.g., Prather, 1996), as a 20% increase in CH₄ abundance requires a 5% smaller emission change than a 20% decrease. Nevertheless, it is clear that the response of surface O₃ to changes in

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global CH₄ abundance is similar to changes in regional NO_x emissions, and we therefore choose to use the same scaling, given in Eq. (4), so that each successive 20 % increase in CH₄ abundance gives a 10 % smaller O₃ increase.

To provide a more critical test of the parameterization, we use it to reconstruct the results of new simulations with the FRSGC/UCI CTM using the RCP scenarios described in Sect. 2. Ozone precursor emission increases are largest in 2030 under the RCP 8.5 scenario, but the RCP 2.6 scenario shows large reductions by 2050, so we explore these two cases which encompass the extremes for the scenarios. A comparison between the monthly mean regional O₃ responses for a full simulation with the FRSGC/UCI CTM against that derived from the parameterization is shown in Fig. 4. In general the parameterization reproduces both the magnitude and the seasonality of the O₃ responses very well. In the RCP 8.5 scenario there are small errors of up to 0.5 ppb in wintertime over East Asia, and this likely reflects increased titration of O₃ associated with the relatively large (65 %) increase in NO_x emissions. Similarly, in the RCP 2.6 scenario the errors are largest in summertime over North America and Europe (reaching 0.8 ppb and 0.5 ppb, respectively), and this is associated with the relatively large NO_x emission reductions (70 % and 50 %, respectively) over these regions. Elsewhere, where emission changes are smaller, the magnitude and the seasonality of the O₃ responses in the full simulations are matched very well, suggesting that the parameterized approach used here is relatively robust.

Figure 4 also presents a regional source attribution for the O₃ changes over each region in each scenario, simplified to show the response to CH₄ abundance changes, regional emissions changes, and emission changes outside the focus region. This source attribution arises naturally from the simple combinatorial approach used here, and provides valuable additional insight without the need for tagged tracers in a full model simulation. The attribution reveals the extent to which the effects of regional emission reductions may be compensated by increases in emissions outside the region and in CH₄, for example over North America in the RCP 8.5 scenario. While there is considerable redistribution of precursor emissions and surface O₃ under this scenario,

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the net change in global surface O_3 due to precursor emission changes is close to zero throughout the year; in effect, the changes in global O_3 are caused almost entirely by the elevated abundance of atmospheric CH_4 under this scenario. The attribution also provides important insight into the role of intercontinental transport in contributing to regional surface O_3 changes, which counteract the effect of regional emission reductions over North America under RCP 8.5, and the effect of regional emission increases over South Asia under RCP 2.6.

Finally, we demonstrate that the approach works well across a range of models. Modelled and estimated surface O_3 responses from four different models simulating the RCP 8.5 2030 scenario are shown in Fig. 5. Each model is treated independently, and the fractional emission changes along this RCP scenario are applied to the O_3 responses from each model. Differences between the models reflect differences in chemistry and transport and in assumptions about the distribution and magnitude of emissions used in the present-day run. The estimates generally lie close to the true response for each model, with both the magnitude and seasonality of the responses reproduced well. The average root mean square (RMS) error in the estimates (0.26 ppb) is much less than the RMS variation between the models (1.2 ppb). We conclude that the approach used here is suitable for estimation of surface O_3 changes across the range of models contributing to HTAP.

5 Anthropogenic contribution to historical trends

To explore the regional anthropogenic contributions to past O_3 trends, monthly mean surface O_3 responses were extracted from each of the 14 models which contributed sufficient results to allow independent parameterization. Where results for global emissions perturbations were unavailable, the responses to rest-of-world perturbations were assumed to equal the ensemble mean responses for the models that did contribute results. The response to changes in tropospheric CH_4 abundance was also included. We use the historical emissions of Lamarque et al. (2010) and derive regional

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and global surface O₃ responses for each model by combining the responses for each precursor over each region based on the fractional emission change from year 2000 emissions. While emission data are available from 1850, we focus on the period from 1960 to present to minimize the errors introduced by applying very large emission reductions. Regional NO_x emissions in 1960 were 40–45 % lower over Europe and North America than in 2000, but were 75–80 % lower over South and East Asia, and the global CH₄ abundance was 30 % lower.

The changes in regional annual mean surface O₃ relative to 2000 conditions are shown in Fig. 6. Regional increases since 1960 vary between 5 and 10 ppb, but the pathways differ substantially, with Europe and North America experiencing increases of about 0.25 ppb yr⁻¹ until 1980, but then smaller increases that turn to a decline in the 1990s, while South and East Asia see steady increases of as much as 0.40 ppb yr⁻¹. The uncertainty in these estimates as represented by the variation between models is relatively small, with a 1σ variation of about ±0.8 ppb since 1960. To test the robustness of these estimates, the FRSGC/UCI CTM was run with 1960 emission changes. The error in estimates for the global and regional annual mean surface O₃ response is typically less than 0.1 ppb, and reaches a maximum of 0.25 ppb for East Asia, where NO_x emissions were 75 % smaller in 1960 than 2000, and thus where we expect to underestimate the O₃ change. Nevertheless, this represents an underestimate of less than 3 % of the calculated change of 8.4 ppb, and we conclude that the parameterization is capable of representing the full model simulations under these conditions.

Figure 6 also shows the contribution from changes in regional emissions, precursor emissions outside the region, and tropospheric CH₄ abundance to the estimated regional O₃ changes on an annual mean basis. For Europe and North America the changes in regional and external precursor emissions have made a similar contribution since 1960, about 2 ppb, although this masks the faster rise and subsequent drop of O₃ from regional sources. Over South and East Asia, regional emission changes contribute 4–6 ppb, more than 50 % of the increase. Increases in atmospheric CH₄ contribute to all regions relatively uniformly, averaging 1.5–1.9 ppb since 1960, and this

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contributes about one third of the O_3 increase seen over Europe and North America. Note the substantial uncertainty in the response to CH_4 reflected in a factor of two difference between the most and least sensitive models, about ± 0.75 ppb since 1960. This reflects differences in chemical environment, particularly in the abundance of OH, as noted elsewhere (e.g., Fiore et al., 2009). Nevertheless, the contribution to the uncertainty in the total O_3 changes since 1960 remains small, only about 0.1 ppb of the 1σ variation, suggesting that models with stronger O_3 responses to CH_4 have weaker responses to other precursors.

These surface O_3 changes due to growth in anthropogenic precursor emissions are compared with observed surface O_3 trends by extracting O_3 responses at sites with long-term measurements, see Fig. 7. The ensemble annual mean surface O_3 matches the observations reasonably well at each location, but the large spread over the different models (as much as ± 16 ppb at Mauna Loa) indicates that this apparent skill is somewhat illusory, hiding differences in annual and diurnal cycles as well as systematic biases due to process representation in the models and to sampling location on the different model grids. The general growth in O_3 is matched well at Mauna Loa, remote from the main source regions considered here, with an average increase in O_3 of about 0.2 ppb yr^{-1} between 1960 and 1990 that levels off over the past decade. The relatively strong interannual variability in the observations is absent from the parameterized trend which uses 2001 meteorology throughout, making direct comparison difficult, but the observed linear trend between 1974 and 2004 is 0.15 ppb yr^{-1} compared with 0.13 ppb yr^{-1} in the models. The contribution of CH_4 increases in the models averages 0.05 ppb yr^{-1} over this period, about 40 % of the trend at this site.

Closer to major continental regions the modelled trends are less well supported by observations. Unfiltered measurements at a coastal site, Mace Head, show an increase of about 0.17 ppb yr^{-1} between 1989 and 2007, and trends in clean baseline air at this site are thought to be as high as 0.31 ppb yr^{-1} (Derwent et al., 2007). Modelled O_3 shows a positive trend of 0.08 ppb yr^{-1} in the 1980s, but the trend turns negative after the 1990s, reflecting reductions in European emissions, and averages

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$-0.03 \text{ ppb yr}^{-1}$ over the measurement period. This may reflect weaknesses in the emission assessments, but it may also reflect the spatial redistribution of emissions within regions which are not represented here (Vautard et al., 2006). Changes in natural sources, compounded by significant interannual meteorological variations, may also contribute to this discrepancy, and the influence of stratospheric O_3 is thought to be significant (Hess and Zbinden, 2011). Further studies at regional scales accounting for spatial emission changes and meteorological variability are required to explain O_3 changes at these continental locations, as previous studies have noted (e.g., Jonson et al., 2006).

6 Application to future trends

We now apply the parametric approach to explore changes in regional surface O_3 along the four RCP emission scenarios. We do not account for any changes in climate, which would differ along the scenarios, but focus on the effects of anthropogenic precursor emission changes alone. Emission changes by 2100 are large along several of the scenarios, so we focus on the period between 2000 and 2050, when changes are smaller and the resulting error in our estimates is less. The ensemble regional mean changes are presented in Fig. 8, and changes by 2050 are summarized in Table 3 along with an estimate of the uncertainty as represented by the 1σ variation over the models. Under all four scenarios surface O_3 falls over Europe and North America, although this fall is reversed over Europe after 2020 along RCP 8.5, driven not by increasing regional emissions but by increasing atmospheric CH_4 . Surface O_3 increases over South Asia in all scenarios, although there is a large difference between RCP 8.5, where increases of more than 5 ppb are seen by 2050, and RCP 6.0, where changes are close to zero. Over East Asia there is also a large variation, with increases in O_3 until 2020 but subsequently decreases under the RCP 2.6 and 4.5 scenarios. It is clear that under all RCP scenarios except RCP 6.0 the greatest increases in surface O_3 are over South Asia, and the large increases here are a major concern given the high population of

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the region and the influence of the region on wider tropospheric composition through strong convective lifting associated with the South Asian monsoon.

For comparison, the O₃ responses along the SRES scenarios used in previous studies are also shown in Fig. 8 and Table 3. The O₃ responses under all four RCP scenarios are substantially smaller than those under the SRES A1B, A2 and B2 scenarios, and all but the RCP 8.5 scenario are smaller than the mildest SRES B1 scenario. This is consistent with the findings of Lamarque et al. (2011) who explored O₃ changes between 2000 and 2100 along the RCP pathways while accounting for changes in climate. Previous assessments of future surface O₃ have typically focussed on the large responses expected under the extreme SRES A2 scenario (e.g., Prather et al., 2003), but the revised assessments of O₃ precursor emissions under the RCP scenarios suggest that the A2 scenario was unduly pessimistic, as earlier studies have noted (Dentener et al., 2006).

The O₃ responses for individual models are shown for RCP 2.6 and RCP 8.5 in Figs. 9 and 10. The responses are broadly similar, although there is a divergence in the results with time as the emissions and CH₄ abundance change. Given that the models differ substantially in their formulation and assumptions, the spread of results provides a simple measure of the uncertainty in O₃ responses. The largest 1 σ variability along the RCP scenarios by 2050 is about ± 0.8 ppb although in a number of cases this is skewed by individual outlying results.

Figures 9 and 10 also provide a clear source attribution of O₃ changes to changes in regional and extra-regional precursor emissions and CH₄ abundance. While the largest contribution to regional O₃ changes under most scenarios is from precursor emissions in the region itself, O₃ transported from sources outside the region can make a substantial contribution that supplements or counteracts these changes. For example, over Europe in 2050 under the RCP 2.6 scenario local emission changes contribute to a reduction of 2 ppb O₃ while changes elsewhere contribute an additional 1.5 ppb reduction. The seasonality of these changes is notably different, as shown earlier in Fig. 4, with regional emission changes having the largest effect in mid-summer when

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photochemistry is most active, and imported O_3 largest in spring. In contrast, over North America in 2020 under the RCP 8.5 scenario the reduction of 2.5 ppb from regional emission reductions is partly offset by a 0.6 ppb increase from emission changes outside the region. This source breakdown provides valuable insight into the potential of regional emission controls in coming decades under these scenarios.

Changes in CH_4 abundance make a major contribution to O_3 changes, particularly for the RCP 8.5 scenario where they increase O_3 2.5–3.0 ppb by 2050, effectively counteracting the benefits of the large precursor emission reductions over North America and Europe. Under this scenario CH_4 abundance reaches 2740 ppb by 2050, a 56% increase over 2000 levels. The difference in CH_4 between the RCP 2.6 and RCP 8.5 scenarios accounts for regional O_3 differences of 3.3–3.9 ppb O_3 by 2050, almost 75% of the O_3 differences between these two scenarios (4.7–5.2 ppb). However, there is substantial uncertainty here as the response to CH_4 differs between models by as much as a factor of two. The parameterization allows us to isolate the contribution of this uncertainty; for the RCP 8.5 scenario it contributes almost half of the 1σ variation in the O_3 response by 2050, 0.2–0.4 ppb. Removing the contribution from CH_4 changes reduces the 1σ variation over Europe from 0.77 to 0.43 ppb and over the global domain from 0.55 to 0.30 ppb. Under the other RCP scenarios CH_4 makes a far smaller contribution to the total uncertainty (typically less than 5%), reflecting both the larger precursor emission reductions in these scenarios and the smaller changes in CH_4 . Nevertheless, it is clear that uncertainty in the response of O_3 to CH_4 can contribute substantially to uncertainty in the estimated O_3 changes, and addressing this discrepancy between models should be a high priority for future model intercomparison studies.

The uncertainty in the O_3 changes estimated here reflects only direct differences in chemical environment in the models under prescribed CH_4 abundances, and CH_4 emissions have not been explicitly considered. In reality, changes in O_3 precursor emissions influence the build-up of CH_4 through control of the OH radical, and thus have an indirect O_3 response through altered CH_4 abundance (Prather, 1996; Wild

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et al., 2001). This long-term O₃ response has been quantified for the HTAP sensitivity studies to derive equilibrium responses (Fiore et al., 2009) but we do not attempt to correct for it under the transient emission scenarios used here. We have also assumed that fractional precursor emission changes can be applied to the present-day emissions in each model and have not attempted to normalise these to some standard values. Previous studies have shown that the effects of an emission perturbation are sensitive to the baseline emissions used (Collins et al., 2008), and this is likely to contribute further uncertainty in the spread of model results. The multi-model studies of Dentener et al. (2006) found global surface O₃ changes between 2000 and 2030 of 1.5 ± 1.2 and -2.3 ± 1.1 ppb under current legislation (CLE) and maximum feasible reduction (MFR) scenarios; we estimate changes of 1.4 ± 0.2 and -2.4 ± 0.5 ppb using the parameterization described here. While the ensemble mean agreement is very good, the variation between models is much less. Although some of this can be attributed to the smaller number of models (14 vs. 26) and the consistency of approach (eliminating differences in scenario interpretation and implementation), it is clear that the 1σ variation calculated here is significantly less than that found in typical model intercomparisons.

The future changes in surface O₃ described here do not include the effects of any changes in climate on atmospheric chemistry or transport. Previous studies have identified changes in continental O₃ associated with faster O₃ production under some climate scenarios, and a consequent increase in the relative importance of O₃ from regional sources over that transported from outside the region (Murazaki and Hess, 2006; Doherty et al., 2011). Other studies have noted that future changes in regional meteorology may alter the build up of surface O₃ (e.g., Mickley et al., 2004). We also neglect the role of meteorological variability in influencing surface O₃ changes (Brown-Steiner and Hess, 2011). Over the large continental-scale regions used here the influence is found to be small (TF-HTAP, 2010; Doherty et al., 2011), but further studies are needed to confirm this.

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7 Applications for policy

The combinatorial approach to estimating surface O_3 responses applied here is easily inverted to provide information on the emission changes required to meet specific O_3 targets over a particular region. This gives valuable insight into the sensitivity of regional surface O_3 changes and provides a useful basis for decisions about emission controls. Figure 11 shows the annual ensemble mean O_3 response in 2050 for each region under each of the RCP scenarios along with its sensitivity to the atmospheric abundance of CH_4 . The close proximity of the O_3 response lines over North America and Europe highlight the similarity in regional emission controls over the four RCP scenarios; for a given CH_4 abundance the O_3 response over Europe between the scenarios differs by only 1.2 ppb. The much wider spread over South and East Asia (more than 4 ppb) reveals the larger differences in regional emissions between the scenarios. The figure provides guidance on the effect of atmospheric CH_4 on regional O_3 . Under the RCP 8.5 scenario, regional surface O_3 over Europe increases by 0.3 ppb between 2000 and 2050; to ensure no increase over this period, the growth of CH_4 would need to be limited to 2610 ppb, a 13 % reduction in the CH_4 increase expected along the RCP 8.5 scenario. Similarly, achieving a 1 ppb decrease in O_3 over Europe by 2050 would require that CH_4 be limited to 2220 ppb, a 47 % reduction in the expected CH_4 increase. Limiting CH_4 to its current atmospheric abundance would lead to a reduction of 2.4 ppb over Europe under RCP 8.5. As the effect of CH_4 changes on O_3 is global, these reductions in the abundance of CH_4 lead to O_3 decreases of a similar magnitude over the other regions considered here.

The effect of regional NO_x emission controls are illustrated in Fig. 12; the CH_4 abundance and emissions of CO and VOC are assumed not to vary in this example. The O_3 responses are truncated for regional emission reductions greater than 80 % where the effect of the reductions is likely to be larger than estimated here due to greater non-linear behaviour. The gradients of the O_3 responses are greater over Europe than over North America, suggesting that a larger fractional NO_x emission change is required

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to give a particular O₃ increment. This may reflect the larger absolute emissions over North America, but may also reveal the greater importance of wintertime O₃ titration over Europe. The curves provide guidance on the likely O₃ benefits from NO_x emission reductions. For example, under RCP 8.5 a 38 % reduction in regional NO_x emissions would be required to maintain European O₃ in 2050 at 2000 levels, greater than the 32 % reduction anticipated for the scenario. A 1 ppb O₃ reduction would require an emission reduction of about 57 %, and a 2 ppb reduction would require an emission reduction of 75 %. Clearly, meeting more stringent O₃ targets than this would require intraregional cooperation focussing on reducing CH₄ or on emission reductions in the developing world, otherwise reductions of more than a few ppb are not possible from controlling European NO_x emissions alone.

8 Conclusions

This study describes a simple parameterization for estimating regional surface O₃ changes based on changes in regional anthropogenic emissions of NO_x, CO and VOCs and in global CH₄ abundance using results from 14 independent global chemical transport models that contributed results to the HTAP model intercomparison. The approach successfully reproduces regional O₃ changes through the year compared with full model simulations from a range of different models under conditions where precursor emissions do not deviate too greatly (typically ±60 %) from those of the present day. While not replacing the need for full model simulations, the approach allows the effects of different emission scenarios to be explored and thus allows identification of scenarios of particular interest for further study. It naturally provides a regional source attribution for surface O₃ changes without the need for tagging tracers in a full model simulation. An additional benefit is that the spread over the ensemble of model results provides a simple measure of uncertainty in regional O₃ responses and their attribution. While the approach does not provide a rigorous quantification of process uncertainty, it allows identification of conditions and regions in which the results of current models

differ substantially. The most important example examined here is that of atmospheric CH₄, where the O₃ response differs by more than a factor of two between models, and this makes the largest contribution to uncertainty in modelled surface O₃ responses for scenarios where CH₄ changes are large (e.g., RCP 8.5).

Application of the approach to historic anthropogenic emission trends captures some of the increase in observed surface O₃ over the past 3–4 decades, but underestimates the magnitude of the O₃ increases observed at continental sites. Previous studies have found similar results (e.g., Lamarque et al., 2010), suggesting that natural sources and changes in climate may have contributed significantly to surface O₃ change. However, the approach is intended for continental-scale use and is not well suited for analysis of observations at particular locations as evolution of the regional distribution of emissions is not accounted for.

Application of the approach to future emission trends following the RCP scenarios demonstrates that substantial annual mean surface O₃ reductions are expected by 2050 over most regions and scenarios, with the exception of South Asia where increases may be as large as 5 ppb. These O₃ responses are contrasted with those from the SRES emission scenarios which show dramatic future increases in surface O₃ driven by large increases in O₃ precursors, consistent with the findings of Lamarque et al. (2011) with a coupled chemistry-climate model. This demonstrates that recent efforts to control precursor emissions are likely to have substantial benefits for future surface O₃ if they are continued into the future.

Much of the difference between the extreme scenarios RCP 2.6 and RCP 8.5 is driven by differences in CH₄ abundance. The importance of CH₄ emission controls for influencing surface O₃ has been highlighted by previous studies (e.g., Fiore et al., 2008), and the present study demonstrates the future importance of this, particularly for RCP 8.5. It also highlights that the O₃ response to CH₄ changes is a major area of uncertainty in current models, and that addressing this will significantly improve estimation of future O₃ changes.

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The uncertainty in future O₃ changes represented here by the spread in results from different models reflects differences in transport and chemical environment in the models, but only captures part of the variation seen in previous model intercomparison studies. A more complete assessment of the uncertainty would require CH₄ emissions to be considered and thus much longer, transient model simulations. Multi-year runs are also required to investigate the effects of interannual variability, and to quantify the effects of climate change on surface O₃ responses, neither of which are considered here. Nevertheless, the approach used provides a good indication of where the largest uncertainties occur. The parameterization used here could be improved by conducting a further set of 20 runs with each of the contributing models that involve complete removal of anthropogenic emissions over each region. This would allow a more robust assessment of the nonlinear behaviour at large emission reductions and thus extend the range of applications. However, given the simplicity of the approach described here and the clear need for further improvement in the models, these refinements are not currently warranted.

Appendix A**Parameterization example**

Ensemble mean annual surface O₃ responses to 20 % regional emission changes for the models used in this study are given in Table 4. Regional emission changes for 2050 from the RCP 8.5 scenario are shown in Table 5; applying these in Eq. (1) gives the mean O₃ responses shown, and matches the results shown in Table 3. Note that the analysis presented in this paper uses monthly mean O₃ responses to follow the seasonal cycle (e.g., see Fig. 5) and treats individual models separately so that the different sensitivities to wintertime titration can be accounted for appropriately. However, the influence of these conditions is relatively small, and the ensemble annual mean response can be estimated to better than 0.05 ppb using the data in Table 4.

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Table 1. Standard and additional simulations for the HTAP model intercomparison showing number of models contributing results for O₃.

Perturbation	None	NO _x	VOC	CO	All
Control run	24				
Methane run	18				
North America		18	16	15	19
Europe		18	16	15	19
South Asia		18	13	14	19
East Asia		18	15	15	20
Global		9	9	9	10

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Table 2. Chemistry transport models used to generate parameterization.

CAM-CHEM	GEM-AQ	INCA-LMDz	MOZECH	TM5-JRC
EMEP	GISS-PUCCINI	LLNL-IMPACT	STOC-HadAM3	UM-CAM
FRSGC/UCI	GMI	MOZART-GFDL	STOCHEM	

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Table 3. Annual regional surface O₃ changes (in ppb) by 2050 along the RCP and SRES scenarios showing model ensemble mean and one standard deviation

Scenario	N. America	Europe	S. Asia	E. Asia	Global
RCP 2.6	-5.6±0.8	-4.7±0.7	0.2±0.8	-3.8±0.5	-2.0±0.5
RCP 4.5	-3.9±0.8	-2.7±0.5	2.9±0.8	-2.5±0.4	-0.8±0.4
RCP 6.0	-2.4±0.7	-2.0±0.5	0.0±0.3	1.4±0.4	-0.4±0.2
RCP 8.5	-0.9±0.9	0.3±0.8	5.2±0.8	1.4±0.6	1.5±0.5
SRES scenarios for comparison:					
SRES B1	-2.0±0.6	-1.2±0.5	2.9±0.6	0.8±0.4	0.8±0.3
SRES B2	5.3±0.5	6.2±1.0	9.2±0.7	7.3±0.7	4.3±0.5
SRES A1B	3.3±0.7	4.6±0.9	10.3±1.0	8.2±1.1	4.5±0.4
SRES A2	6.9±0.7	7.7±1.3	11.7±0.8	9.1±0.8	6.2±0.7

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Table 4. Model ensemble mean annual surface O₃ changes (in ppb) used in the present analysis.

Model Scenario		N. America	Europe	S. Asia	E. Asia	Global
SR1	Control run	36.125	37.816	39.562	35.614	27.226
Global abundance reduction						
SR2	-20 % global CH ₄	-1.111	-1.195	-1.244	-1.045	-0.906
North American emission reductions						
SR3NA	-20 % NO _x emissions	-0.749	-0.212	-0.104	-0.125	-0.127
SR4NA	-20 % VOC emissions	-0.285	-0.108	-0.046	-0.065	-0.059
SR5NA	-20 % CO emissions	-0.104	-0.061	-0.035	-0.038	-0.027
European emission reductions						
SR3EU	-20 % NO _x emissions	-0.075	-0.445	-0.147	-0.115	-0.073
SR4EU	-20 % VOC emissions	-0.087	-0.448	-0.090	-0.115	-0.078
SR5EU	-20 % CO emissions	-0.030	-0.113	-0.033	-0.037	-0.021
South Asian emission reductions						
SR3SA	-20 % NO _x emissions	-0.037	-0.039	-1.074	-0.093	-0.064
SR4SA	-20 % VOC emissions	-0.020	-0.021	-0.195	-0.029	-0.020
SR5SA	-20 % CO emissions	-0.019	-0.019	-0.097	-0.025	-0.015
East Asian emission reductions						
SR3EA	-20 % NO _x emissions	-0.112	-0.076	-0.091	-0.592	-0.083
SR4EA	-20 % VOC emissions	-0.070	-0.057	-0.045	-0.306	-0.050
SR5EA	-20 % CO emissions	-0.045	-0.046	-0.041	-0.126	-0.028
Rest-of-world emission reductions						
SR3RW	-20 % NO _x emissions	-0.125	-0.123	-0.182	-0.164	-0.290
SR4RW	-20 % VOC emissions	-0.038	-0.043	-0.044	-0.055	-0.051
SR5RW	-20 % CO emissions	-0.035	-0.034	-0.050	-0.040	-0.038

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Table 5. Regional emission changes and estimated O₃ responses for 2050 from the RCP 8.5 scenario.

Applied change	N. America	Europe	S. Asia	E. Asia	Rest-of-World	Global
CH ₄ abundance						56.5 %
NO _x emissions	−52.4 %	−32.1 %	61.3 %	−9.4 %	7.1 %	
VOC emissions	−57.2 %	−19.0 %	61.0 %	5.8 %	2.1 %	
CO emissions	−70.7 %	−50.7 %	34.5 %	−20.6 %	−5.7 %	
Resultant annual mean regional surface O ₃ responses (ppb)						
O ₃ response	−0.91	0.32	5.25	1.42		1.49

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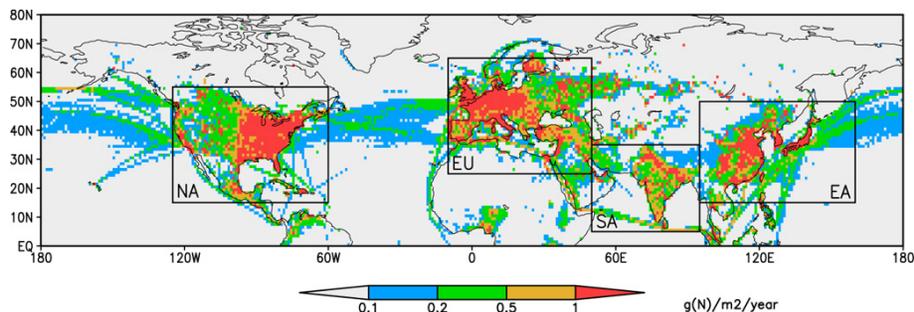


Fig. 1. Anthropogenic surface NO_x emissions ($\text{g N m}^{-2} \text{yr}^{-1}$) for year 2000 from Lamarque et al. (2010) showing the HTAP source-receptor regions considered here: N. America, Europe, S. Asia and E. Asia.

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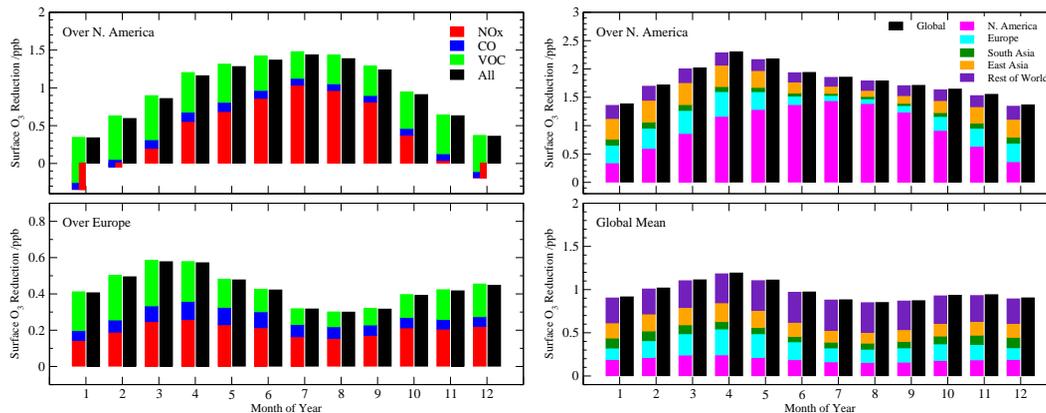


Fig. 2. Tests of linearity with the FRSGC/UCI CTM. **(a)** Sum of O₃ responses from 20% reductions in anthropogenic NO_x, CO and VOC emissions from North American sources compared with responses from a combined 20% emissions change, shown over the source region (upper panel) and a distant receptor region (Europe, lower panel). The contribution from NO_x is negative over the source region in winter, partly compensating for the effects of CO and VOC emissions. **(b)** Sum of O₃ responses from combined 20% emission reductions over each of the HTAP regions and over the rest of the world compared with the response from a global emission change, shown over a single region (N. America, upper panel) and over the globe (lower panel).

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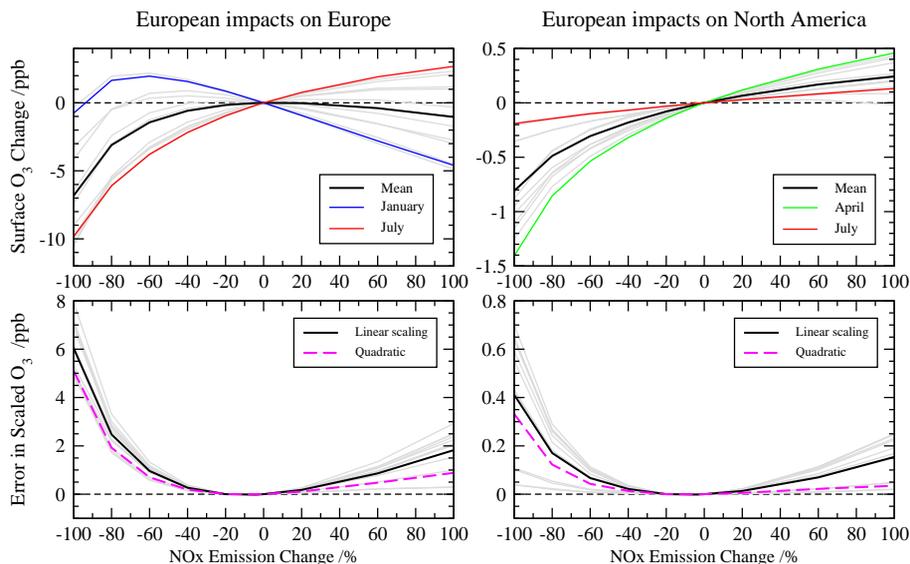


Fig. 3. Sensitivity of monthly O_3 changes in the FRSGC/UCI CTM to the magnitude of European NO_x emissions relative to current conditions over the source region (left) and a downwind receptor region (N. America, right). Annual mean O_3 responses are shown in black and individual months in grey; months with the largest and smallest responses are highlighted. The lower panels show the error associated with linearly scaling a 20% emission perturbation over the source and receptor regions (black) and using a quadratic scaling (magenta).

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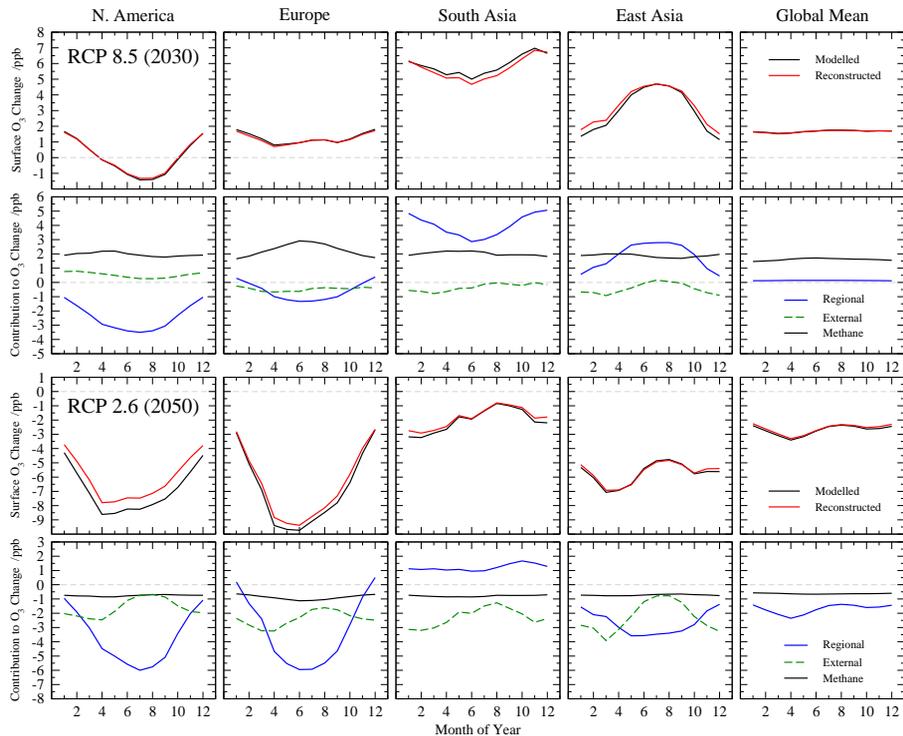



Fig. 4. Monthly regional mean surface O₃ changes in the FRSGC/UCI CTM between 2000 and 2030 for the RCP 8.5 emissions (upper panel) and between 2000 and 2050 for the RCP 2.6 emissions (lower panel). The upper row of each panel shows the estimated O₃ response compared with that from a full simulation, and the lower row shows the attribution of the O₃ response to global CH₄ changes (black) and to emission changes inside (blue) and outside (green) the source region.

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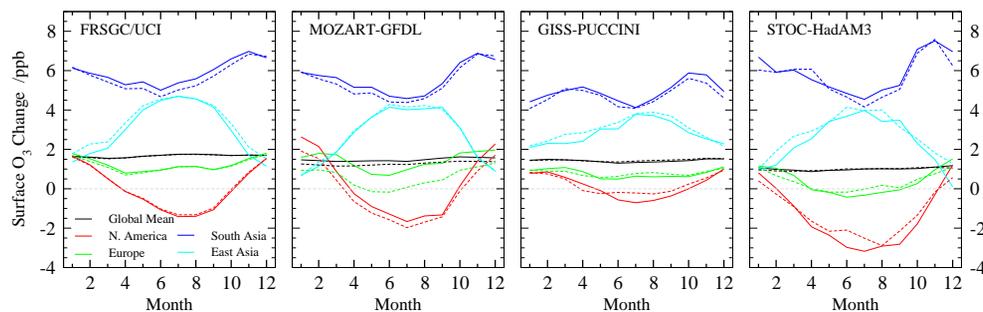


Fig. 5. Monthly regional mean surface O_3 changes between 2000 and 2030 following the RCP 8.5 scenario with the FRSGC/UCI CTM, MOZART-GFDL CTM, GISS-PUCCINI GCM, and STOC-HadAM3 GCM (solid lines) and the parameterized estimate for each model (dashed lines).

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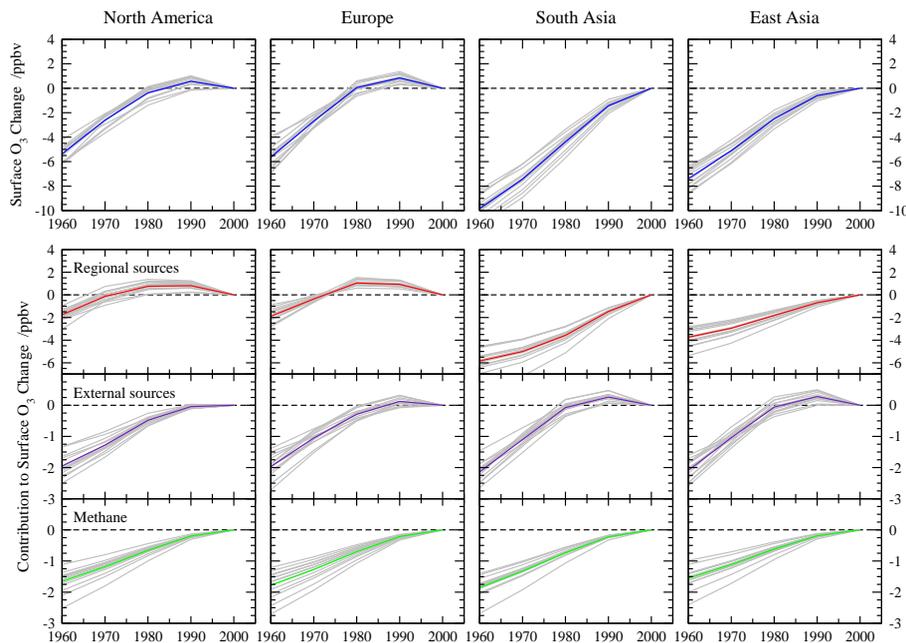


Fig. 6. Annual regional mean surface O_3 changes relative to 2000 over each HTAP region following historical precursor emission changes between 1960 and 2000 (top row), and the contribution of regional anthropogenic sources, anthropogenic sources outside the region, and global methane changes (bottom row). Individual model responses are shown in grey and the mean of all 14 models is coloured.

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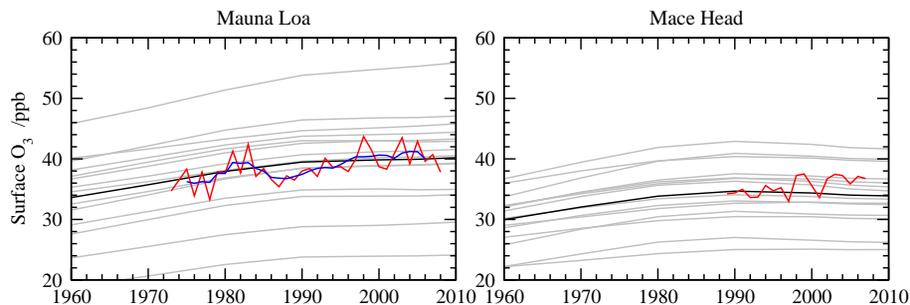


Fig. 7. Annual mean surface O_3 trends at Mauna Loa and Mace Head between 1960 and 2010 for individual models (grey) and ensemble mean (black) against annual mean observed surface O_3 (red). The 5-year running mean of the observations at Mauna Loa is shown in blue.

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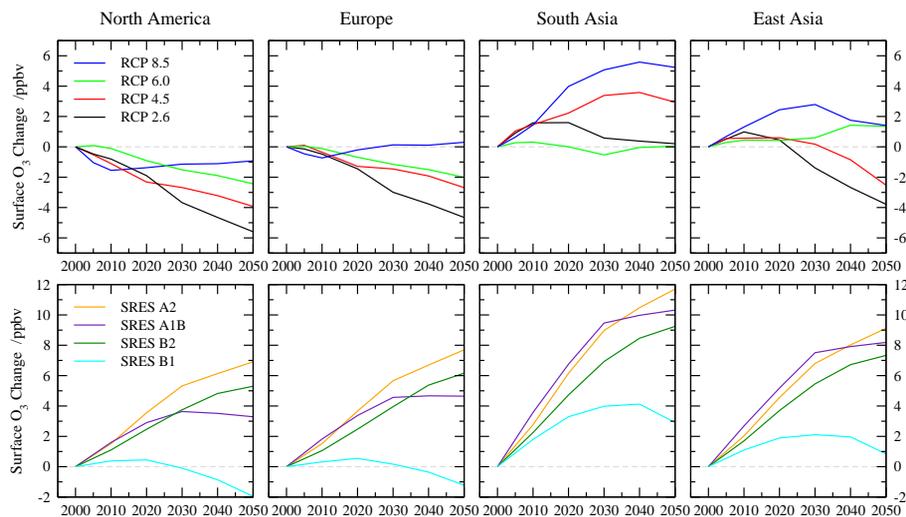


Fig. 8. Model ensemble annual regional mean surface O_3 changes over the four HTAP regions from the parameterization following (a) the different RCP precursor emission pathways and (b) the SRES scenarios. The y-axis spans an O_3 change of 14 ppb in each case to allow direct comparison of the magnitude of O_3 changes.

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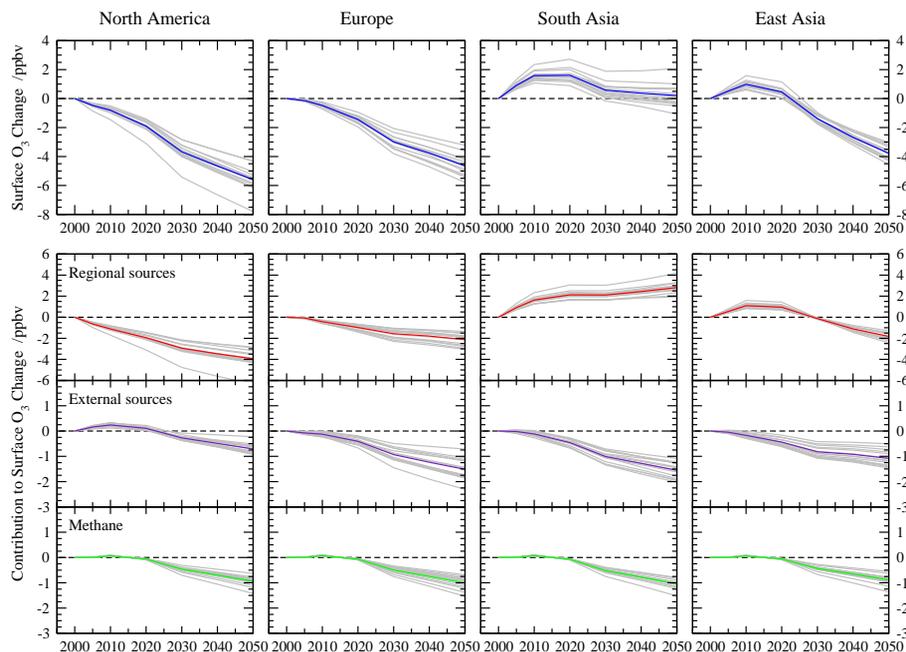


Fig. 9. Regional mean O_3 changes along the RCP 2.6 scenario showing the ensemble mean (coloured line) and individual model responses (grey lines). A source attribution is presented for each region in the lower panels.

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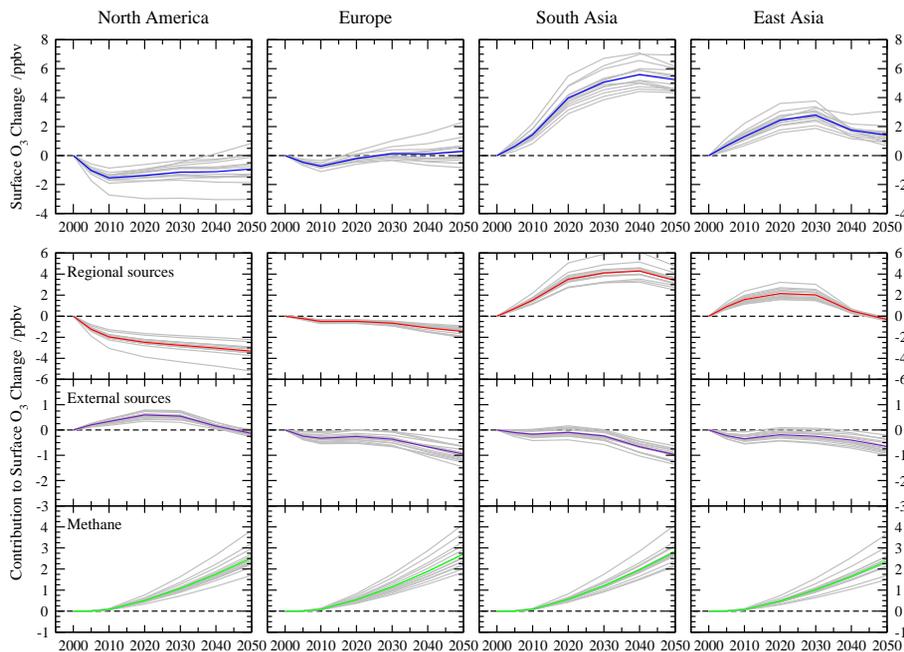


Fig. 10. As Fig. 9 for the RCP 8.5 scenario.

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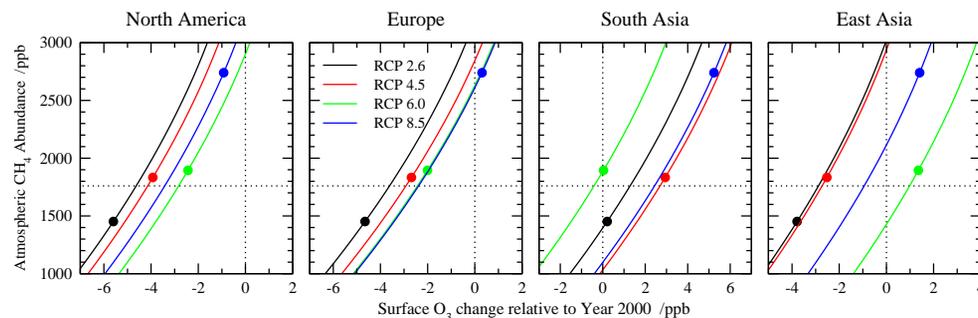


Fig. 11. Sensitivity of regional surface O₃ in 2050 to the atmospheric CH₄ abundance under each of the RCP scenarios. Circles mark the ensemble mean surface O₃ response under each scenario, and curves show how this would change under different levels of CH₄. Dashed lines mark CH₄ and surface O₃ for year 2000.

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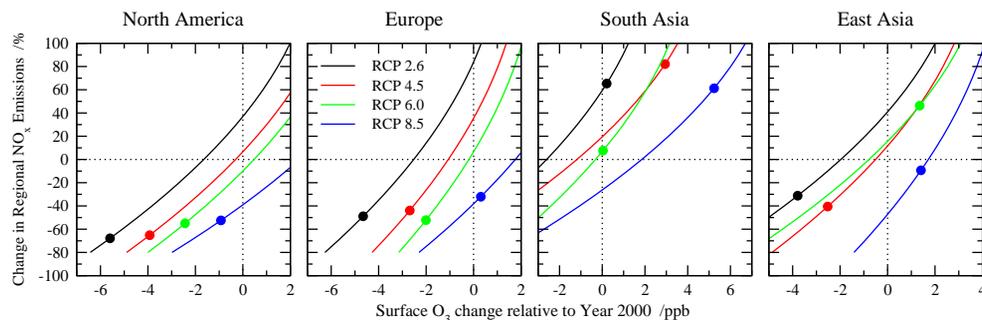


Fig. 12. Sensitivity of regional surface O₃ in 2050 to regional NO_x emissions under each of the RCP scenarios. Circles mark the ensemble mean surface O₃ response under each scenario, and curves show how this would change for different regional NO_x emissions. Dashed lines mark year 2000 conditions.

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